Nondispersive two-electron Trojan wave packets

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We demonstrate the existence of stable non-dispersing two-electron Trojan-like wave packets in the helium atom in combined magnetic and circularly polarized microwave fields. These packets follow circular orbits and we show that they can also exist in quantum dots. Classically the two electrons follow trajectories which resemble orbits discovered by Langmuir and which were used in attempts at a Bohr-like quantization of the helium atom. Eigenvalues of a generalized Hessian matrix are computed to investigate the classical stability of these states. Diffusion Monte Carlo simulations demonstrate the quantum stability of these two-electron wave packets in the helium atom and quantum dot helium with an impurity center.

PACS numbers: 31.50.+w, 32.80.Rm, 42.50.Hz, 95.10.Ce

The direct manipulation of atoms and ions at the quantum level is currently a flourishing area of physics [1]. For example, ion traps combined with laser cooling techniques have been used to create new states of matter including ion liquids, Wigner crystals and Bose-Einstein condensates [2, 3]. The ability to manipulate the quantum properties of matter directly, as exemplified by these advances, is central to the practical development of nanodevices, e.g., quantum dots and microchip traps [4].

Recently it has proved possible to create a trap consisting of a single atom inside of which the quantum behavior of an electron can be manipulated [5]. In these experiments [5, 6] an electron in an excited lithium atom was localized in a classical orbit almost indefinitely, neither spreading nor dispersing. This "classical atom" was synthesized by "tethering" the electron using a microwave field to which its motion is phase locked. Potential practical applications of the technique include Rydberg tagging in molecular spectroscopy and the preparation of stable antimatter atoms [6].

Although only recently realized experimentally, the existence of coherent, non-dispersive one-electron wave packets in Rydberg atoms was predicted about a decade ago using essentially classical mechanical arguments [7, 8, 9, 11]. A peculiar property of this type of wave packet is that the electron is localized at an equilibrium which corresponds to an energy *maximum* in the noninertial frame. Such equilibria are similar to the well known Lagrange equilibrium points L_4 and L_5 in the restricted three-body problem of celestial mechanics at which, e.g., Jupiter's Trojan asteroids are located [10, 11]. Because their classical dynamical origins are so similar, these wave packets have come to be known as Trojan wave packets [7, 11].

Naturally it is interesting to ask if similar wave packets but involving more than a single electron can be synthesized. In fact, considerable effort has already been directed towards this goal. For example, two-electron wave packets in barium atoms [12] have been prepared and provide a powerful tool in the study of electron correlation in atoms [13]. However, while these wave packets are prepared initially in localized radial states using laser pulses they eventually disperse, e.g., through a collision near the nucleus. In another approach a particular type of dynamical stabilization is used to generate nondispersing wave packets in two-electron atoms. In this case it is necessary that one of the electrons is well localized relatively close to the nucleus [13].

Here we report the existence of coherent, two-electron, non-dispersing wave packets in helium-like atoms (or quantum dot helium) which are true analogs of the one-electron Trojan wave packets; both electrons follow classical orbits in direct analogy with the states prepared experimentally in Refs. [5, 6]. Unlike the one-electron Trojan problem, this system is a genuine quantum three-body problem and thus represents a more direct analogy with the classical restricted three-body problem. Stable two-electron equilibria are produced through the simultaneous application of combined circularly polarized (C.P.) electromagnetic and magnetic fields to the helium atom or to quantum dot helium with an impurity center [14]. The equilibria so produced are stable over broad ranges of field parameters and the wave packets are, therefore true non-dispersing coherent-like states.

We demonstrate that, in the two-dimensional limit, e.g., as in quantum dot helium, it is possible to create a variety of stable two-electron, non-dispersive, Trojan wave packets. However, there exists only a single stable three-dimensional configuration in the helium atom itself. These latter wave packets are actually two-electron examples of the electronic coherent states sought after by Schrödinger [15] in the hydrogen atom. In particular these wave packets follow the "double-circle" orbits originally discovered by Langmuir [16] in the helium atom. We also present diffusion Monte Carlo simulations which confirm directly the quantum stability of these states. Further, we show that it should be possible to produce certain of these states directly in quantum dots using field strengths which are currently accessible in the laboratory.

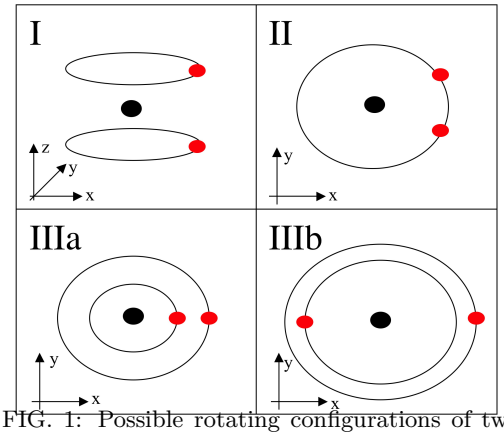

FIG. 1: Possible rotating configurations of two electrons in a helium atom or quantum-dot helium in combined magnetic and CP fields. Type I: Langmuir configuration, type II: transverse configuration, type III: collinear configurations (a and b).

In a coordinate system rotating with the C.P. field and assuming an infinite nuclear mass the Hamiltonian for the helium atom interacting with a C.P. field and a magnetic field perpendicular to the plane of polarization is, in atomic units,

$$H = H_1 + H_2 + \frac{1}{r_{12}} \tag{1}$$

where,

$$\begin{aligned} H_i &= \frac{\mathbf{p}_i{}^2}{2} - \frac{2}{r_i} - (\omega \pm \frac{1}{2})(x_i p_{y_i} - y_i p_{x_i}) \\ &+ \frac{1}{8}(x_i^2 + y_i^2) + \mathcal{E} x_i \end{aligned} \tag{2}$$

with $i = 1, 2$; $r_i = \sqrt{x_i^2 + y_i^2 + z_i^2}$ and $\mathbf{p}_i = (p_{x_i}, p_{y_i}, p_{z_i})$ are the coordinate and momentum vectors of each electron; ω and $\mathcal{E}$ are the scaled C.P. field frequency and scaled field strength, respectively [17]; the cyclotron frequency in scaled units is ± 1. With a re-interpretation and re-scaling of parameters this is also the Hamiltonian of quantum dot helium with an off-axial impurity centre in a magnetic field [18]. Equilibria of the classical Hamiltonian can most easily be found by constructing a zero-velocity surface (ZVS) [10], which shares some properties with a potential energy surface (PES) but contains addition terms due to the centrifugal and Lorentz forces [10, 11]. An extended discussion of the construction of the ZVS in atomic systems is given in [11, 19] - briefly the ZVS is obtained by re-writing the Hamiltonian in terms of velocities rather than momenta. This gives

$$H_i = \sum_k \frac{1}{2}\dot{q}_k^2 - \frac{1}{r_i} + \mathcal{E} x_i - \omega(\omega \pm \frac{1}{2})(x_i{}^2 + y_i{}^2) \tag{3}$$

where $q_k = x, y, z; k = 1, 2, 3$. Setting the velocities to zero produces the ZVS which, in the present case, has the form

$$V_{ZVS} = V_{ZVS_1} + V_{ZVS_2} + 1/r_{12} \tag{4}$$

$$V_{ZVS_i} = -\frac{1}{r_i} + \mathcal{E} x_i - \omega(\omega \pm \frac{1}{2})(x_i{}^2 + y_i{}^2) \tag{5}$$

The ZVS reduces to a true PES when $\omega = 1/2$, i.e., when the paramagnetic term in eq. (2) vanishes. As with a PES equilibria of the motion are then obtained as extrema of the ZVS (maxima, minima and saddles) [11, 19].

We find three possible types of equilibrium point whose stability depends on the particular values of the parameters. Type I: This configuration, illustrated in Fig. 1 (I), corresponds to the unstable "two-circle" orbits discovered by Langmuir [16] in which the (classical) electrons occupy parallel orbits located above and below the plane of the nucleus with longitudes of 30^o. Stabilization is possible only through the application of external fields. Type II: Illustrated in Fig. 1 (II), this configuration has the two electrons orbiting in the same orbit in a plane containing the nucleus. The angle subtended by the electrons at the nucleus can be arbitrary. This configuration can only be stable in two spatial dimensions e.g., a quantum dot. Type III: This configuration, of which two variants are possible - IIIa and IIIb in Fig. 1 - is unstable in the helium atom itself but stable in 2-dimensional quantum dot helium where it appears as variant IIIa. Both electrons lie in the plane of polarization and, remarkably, lie on the same side of the nucleus.

The equilibrium corresponding to the Langmuir configuration has the geometry of an equilateral triangle; i.e., the three particles form an equilateral triangle whose sides are of length a (see Fig. 1 a and Fig. 2). The relationship between the field parameters and the size of this triangle is defined by the following cubic in a;

$$(\omega^2 \pm \omega)\frac{a^3}{2} + \frac{\sqrt{3}}{3}\mathcal{E} a^2 - 1 = 0 \tag{6}$$

Note that the presence of external fields does not change the geometry of the Langmuir configuration. Figure 2 shows the dependence of a on the electric field strength for selected frequencies ω. We perform an extended stability analysis of the system around these trajectories, i.e. we study small oscillations [20], in the rotating frame. The generalized Hessian matrix (stability or monodromy matrix) for a system of n-particles in the rotating frame can be written as

$$\mathbf{S} = \mathbf{H}_e \mathbf{R} \tag{7}$$

where

$$\mathbf{R}_{a_i b_i} = sgn(a_i)\delta_{a_i, b_i} \tag{8}$$

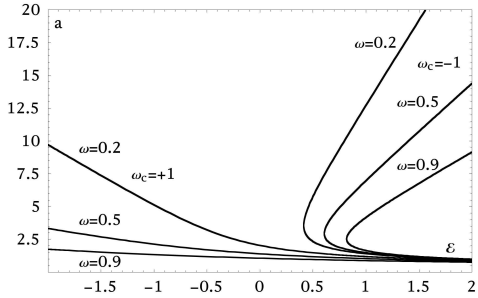

FIG. 2: Dependence of the length of the sides of the equilateral triangle described by the three charges in the rotating frame on the strength of the CP field for various rotation frequencies. Note that for $\omega_c = -1$ the system is bistable and only the larger solution corresponds to the stable trajectory. When $\omega_c = -1$ there is no critical field and only a single solution [17].

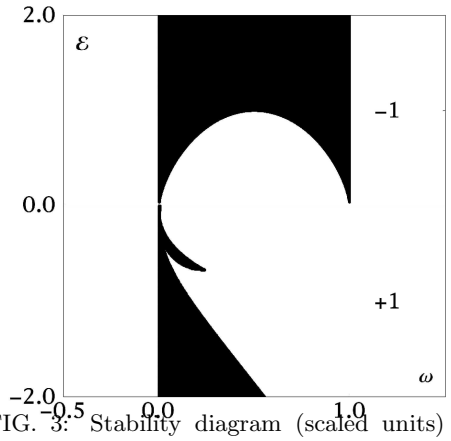

FIG. 3: Stability diagram (scaled units) of the disturbed Langmuir trajectory for anti-centrifugal Lorentz force (upper half, $\omega_c = -1$) and co-centrifugal (lower half, $\omega_c = +1$) as a function of scaled rotation frequency and the electric field. The black regions corresponds to stable trajectories. Note that no magnetic field $\omega_c < \omega$ can lead to stability in case -1 and magnetic field weaker than rotation $\omega_c < \omega$ is permitted to obtain stability in case $+1$. This suggests a maximum of the ZVS and Paul trap-like (Trojan) stabilization.

where $sgn(a_i) = 1, a_i = p_i$, $sgn(a_i) = -1, a_i = x_i$ and H_e is the phase space defined Hessian matrix

$$(\mathbf{H}_e)_{a_i b_i} = \frac{\partial^2 H_{osc}}{\partial a_i \partial b_i} \tag{9}$$

$a_i = q_i, p_i$, $b_i = q_i, p_i$ and H_{osc} is the harmonic Hamiltonian around the orbit [7, 21].

The generalized Hessian matrix can be written explicitly as

$$\mathbf{S}_{ij} = \begin{bmatrix} \mathbf{A} & \mathbf{0} & \mathbf{C}_{11} & \mathbf{C}_{12} \\ \mathbf{0} & \mathbf{A} & \mathbf{C}_{21} & \mathbf{C}_{22} \\ \mathbf{B} & \mathbf{0} & \mathbf{A} & \mathbf{0} \\ \mathbf{0} & \mathbf{B} & \mathbf{0} & \mathbf{A} \end{bmatrix} \tag{10}$$

Note that the right upper block is the normal Hessian matrix of the stationary mechanical system C_{ij} [20] and

$$\mathbf{A} = \begin{bmatrix} 0 & \omega \pm \frac{1}{2} & 0 \\ -\omega \mp \frac{1}{2} & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \mathbf{B} = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix} \tag{11}$$

Figure 3 shows the stability island as a function of scaled coordinates [8] for the cases $\omega_c = 1$ (lower half) and $\omega_c = -1$. Remarkably, unlike for Trojan wavepackets both the circularly polarized field and the magnetic field are necessary to stabilize the Langmuir trajectories. One may notice an interesting v-shaped valley which is similar to the "negative mass" case of Ref. [22].

The quantum wave function for the monodromy matrix for the stable trajectory [21] may be written [7]

$$\psi = e^{-\sum_{ij} A_{ij} x_i x_j} \tag{12}$$

where A_{ii} are always real and A_{ij}, $i \neq j$ imaginary to guarantee normalizability of the quantum wavefunction in the rotating frame. Because the wavefunction is localized around the stable point in the configuration plane the contraction (integration) of diagonal of the density matrix $\psi^*(x)\psi(x)$ over a single electron variable leads to the single electron density localized around electron equilibria and corresponds to a nondispersing two-electron wavepacket moving in the laboratory frame. The detailed calculations are quite cumbersome and will be given elsewhere. As one can see from Fig. 3 the system never stabilizes for magnetic fields weaker than the electric field frequency for the case of anti-centrifugal Lorentz force and may stabilize for such fields for faster rotations for the co-centrifugal Lorentz force.

In order to check for the existence of the quantum states themselves we solved the time dependent Schrödinger equation using the Diffusion Monte Carlo method [23, 24]. Fig. 4 shows density plots of the wave function of each electron. The probability density is clearly localized around the equilibrium points. The wavepackets are actually approximate eigenstates of the Hamiltonian in the rotating frame and, therefore, do not disperse in the inertial frame. These eigenstates are localized around a classical equilibrium point whose local spectrum is an almost-harmonic ladder of coherent states. That is, the eigenstate of the atom in the fields itself behaves as a wavepacket evolving according to the classical equations of motion. For the large field values

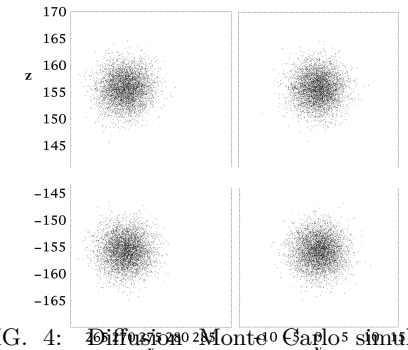

FIG. 4: Diffusion Monte Carlo simulations of Langmuir wavepackets in the rotating frame for $\Omega_c = 0.0370$ a.u., $\mathcal{E} = 0.1235$a.u., and $\Omega = \Omega_c/2$. The density of points reflects the wavefunction itself not probability density [18, 23]. The wavepackets are approximate eigenstates of the Hamiltonian in the rotating frame and follow a circular orbit in the inertial frame.

employed here the Langmuir wavepackets actually correspond to the ground state of eq. (2). This is similar to the case of the original Trojan wavepackets [7] which are the ground states of the locally harmonic Hamiltonian obtained by expansion around the Trojan equilibria which are, in that case, energy maxima [8]. We neglect finite nucleus mass effects here but on the timescales of interest, as in the case of one-electron Trojan wavepackets, these effects will be negligible [25].

In conclusion we have demonstrated that Langmuir trajectories in magnetic and circularly polarized fields are stabilizing for certain parameter regions when the magnetic field is parallel to the rotation axis and the C.P. field is perpendicular. Corresponding quantum states exist. This is also true for two-dimensional quantum dot helium. These regions are not possible to predict from purely analytical considerations and require extensive numerical searches in parameter space. For example, for a quantum dot of radius 100 nm (Type IIIa configuration) the equilibria are at 62.72 nm and 98.00 nm from the nucleus. To achieve this configuration a magnetic field of 5 T is applied and the impurity with the effective charge Z_{eff}=0.008 e displaced from the center of the parabolic dot by 98 nm generates effective "microwave" field of frequency 548 GHz and strength 4.593 kV/m.

Financial Support by the (US) National Science foundation and the Petroleum Research Fund, administered by the American Chemical Society is gratefully acknowledged.

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